The characteristics of environmental particulate matter in the urban area of Beijing, China, during the 2008 Olympic Games

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Abstract

Atmospheric particulate matter (PM) and street dust samples from the Chaoyang District of eastern Beijing were studied over a period encompassing the 2008 Beijing Olympic Games. PM_{10} concentration data are combined with trajectory clustering and potential source contribution function (PSCF) methods to identify the principal transport pathways. Sources for high-concentration aerosol events and airflow from the surrounding Hebei Province and Shandong Province to the southeast are found to exert the most significant external influence on Beijing's air quality. China undertook a number of initiatives to improve air quality for the Olympic Games and we show that PM_{10} concentrations and magnetic susceptibility were significantly lower during the Olympic period compared to the pre-Olympic period confirming that controlling local sources in Beijing and shutting factories in surrounding provinces substantially improved air quality. On short timescales PM_{10} shows an inverse correlation to relative humidity and hence precipitation which acts to improve air quality. Atmospheric PM and street dust remained high through the Olympic period probably due in part to redistribution of historical sources and implying that the aim of zero pollution is not achievable in the short term. Analysis of the heavy metal content in both PM and street dust identifies consistently high values of Zn, with Pb relatively higher in the PM; a primary source in vehicular emissions therefore seems likely.

Keywords: Atmospheric pollution; 2008 Olympic Games; Beijing; PM_{10}; magnetic susceptibility; heavy metals

1. Introduction

Particulate matter (PM) in the atmosphere and in ground deposits can originate from natural sources (dust blown into the air by wind, salts splashed into the air by sea spray and soot from volcanoes and forest fires) and from various anthropogenic activities of which the biggest sources are vehicle and smokestack emissions, and the creation of dust generated when vegetation has been removed for construction or grazing purposes. When averaged globally, anthropogenic PM appears to account for about 10% of the total aerosol amount (Perrino, 2010) but this figure varies greatly from place to place as do the chemical compositions and inferred sources. PM can vary in size from sub-micron aerosols to visible dust particles: the coarse particles rapidly removed from the air by sedimentation are of local impact only, whereas fine particles can have a global reach (Perrino, 2010). Urban surfaces typically receive fine PM issued from remote sources through atmospheric transport as well as a wider range of particle sizes from local human activities (Harrison et al., 1981; Thornton, 1991). Street dust can also be easily re-suspended back into the atmospheric aerosol by wind (Wise and Comrie, 2005) or vehicle movement (Almeidam et al., 2006). Analysis is therefore complex but the integrated data from studies of PM in street dust nevertheless provides the essential basis for understanding atmospheric pollution and assessing effects on human health (e.g. Hien et al., 1999).

PM typically contains magnetic particles characterized by stable and intense magnetic properties (Maher et al., 1999) with this magnetic fraction linked closely to heavy metals such as zinc, cadmium and chrome (Georgeaud et al., 1997) but also to mutagenic organic compounds (Morris et al., 1995), all of which are dangerous to human health. Thus magnetic properties provide valuable proxies for deducing the origin of PM and because of their control by the above factors they become a convenient signature of air pollution (Qiao et al., 2013). Routinely-measureable magnetic parameters provide information on the concentration, domain state (or indirectly the magnetic grain size), and mineralogy of magnetic particles and collectively these are related to original geological or subsequent environmental processes (Liu et al., 2012). Statistical methods such as trajectory clustering have been widely used to identify the pathways and sources of air pollution (e.g. Ashbaugh, 1983; Sirois and Bottenheim, 1995; Wang et al., 2006; Borge et al., 2007). In the present investigation we study magnetic susceptibility in atmospheric fallout samples and apply statistical clustering technique to a 5-month dataset of atmospheric trajectories to identify the particulate matter sources and long-range transport patterns that can influence air pollution.

The 29th Olympic and Para-Olympic Games were held between 2008 August 8 and September 17, in Beijing, a densely populated city with more than 16 million residents and 3 million motor vehicles. Traffic congestion and air pollution thus presented two major challenges to the organizers of the games. To improve air quality and control traffic a series of measures were implemented which included the relocation of industrial plants with large emissions outside of the city and the implementation of new standards to reduce vehicular emission and limit their use. Domestic controls included a progressive switching to clean fuels and low-sulfur coal for household use before and during the Olympic period (Li et al., 2010; Zhou et al., 2010). Whilst most of these measures were intended to have a lasting effect, the vehicular restrictions were largely temporary in nature and are therefore expected to be detectable for only a limited time period. This research evaluating the impact of these air pollution control measures is therefore classified as “before”, “during” and “after” the Games. The present study has had two objectives: firstly we have aimed to determine the source regions influencing the air in Beijing in order that effective source control strategies can be put into place in the longer term; secondly, we have sought to evaluate the relationship between atmospheric PM and street dust. It is well known that the re-suspension of road dust particles from urban street surfaces is an important source of atmospheric PM pollution (Amato et al., 2009; Martuzevicius et al., 2011; Zhao et al., 2016), and the measurement of atmospheric deposits on street surfaces can be useful for studying deposition over a longer time intervals.

2. Experimental Section

2.1. Sample collection

Atmospheric PM was determined by the gravimetric method at monthly intervals from June 2008 to March 2009 in the Chaoyang Distinct of eastern Beijing. The PM samples were collected in 15×30 cm cylindrical glass vessels containing glycol and the vessels placed on a 1.5 m sampling frame. PM samples were collected at two sampling sites: the first was a residential location at Sanlitun (SLT) near the Chaoyang Park (CY) containing the Olympic Site and the second was an industrial site, Fatou (FT) located near the Jing-Shen Highway. The Olympic Park is located at the northwestern sector of Chaoyang district (Fig. 1), and street dust samples were collected on roads with different traffic densities or pavements around the Olympic Park between November 2007 and October 2008. The sampling sites were selected from the traffic avenue and forest park inside the Olympic Park (AT) near to the north 5^{th} ring road (Fig. 1) with samples collected using a nylon brush and non-magnetic scoop from squares 0.5 to 1 m^2 in area prior to transfer to clean, self-sealing polyethylene bags. To evaluate the relationship between PM and street dust over the interval of enforcement measures, the samples were selected for analysis between June 2008 and October 2008. The details of sampling sites and methods are described in Qiao et al. (2011a, b).

2.2. Methods

All samples were air-dried and sieved through a mesh of size 500 μm to remove obvious refuses and the dust residue tightly packed into 10cc polyethylene cubes for magnetic measurements in the Paleomagnetism and Geochronology Laboratory at the Institute of Geology and Geophysics, Chinese Academy of Sciences. A total of 17 cubes from 4 sites were prepared for analysis in this way. Magnetic susceptibility measurements using a Kappabridge KLY-3 were accompanied by determination of Fe, V, Cr, Co, Ni, Cu, Zn and Pb heavy metal contents using inductively-coupled plasma-mass spectrometry (ICP-MS); the latter technique employed the DZ/T0223-2001 method with HR-ICP-MS (Element I) Finning MAT equipment located in the Analytical Laboratory of the Beijing Research Institute of Uranium Geology. The averaged mass amounts used for chemical measurement were 10 mg, and for magnetic measurements were respectively 0.141g (PM samples) and 4.025g (street dust samples). Although the unit for mass specific susceptibility (χ) is more generally preferred, for the very small sample amounts involved with PM samples, the magnetic susceptibility is conveniently denoted by the volume susceptibility value (κ).

The Geo-accumulation index, I_{geo}, is defined as $I_{geo} = \log_2 (C_n/1.5B_n)$ where C_n is the measured concentration of the heavy metals in the environment; B_n is the geochemical background value in soil (CNEMC, China National Environmental Monitoring Center, 1990), and the factor 1.5 is introduced to minimize effects of possible variations in the background values as originally assessed from studies of bottom sediments (Müller, 1969). This index provides a simple factor for assessing the impact of natural geological processes on the natural background values and the influence of human activity responsible for the heavy metal pollution. The geo-accumulation index consists of 7 classes or grades (Table 1), whereby the highest class 6 reflects a 100-fold enrichment above the background values (Forstner et al., 1990).

To accommodate external processes 3-day back-trajectories arriving at Beijing (39.9N, 116.4E) 500m above ground level (a.g.l) were calculated using the Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) model (Stein et al., 2015; Rolph, 2016) loaded into the geographic information system (GIS) based software, TrajStat (Wang et al., 2009). The NCEP (National Centers for Environmental Prediction) archive data downloaded from NOAA provided the meteorological data for input into the model. The PM_{10} concentrations for Beijing from June 2008 to October 2008 were calculated from the air quality index (AQI) reports for major Chinese cities and, for the purposes of statistically analyzing the data, PM_{10} concentrations at Beijing were assigned to corresponding trajectories using the Euclidean distance for the trajectory clusters.

3. Results and discussion

3.1. Transport Pathways and sources

Although PM sources in Beijing clearly contribute to PM concentrations in the city, it has already been shown that surrounding areas contribute to Beijing's PM concentrations (Streets et al., 2007). Five clusters were therefore produced by the clustering algorithm (a total of 152 trajectories classified into 5 clusters) to determine the impact of different source regions on the PM concentration in Beijing; these cluster-mean trajectories are shown in Fig. 2 by different colors. The air masses associated with clusters 1 and 2 have traveled over desert and semi-desert regions of Inner Mongolia on the way to Beijing; cluster 3 air is anticipated to have initially followed paths over Russia subsequent to passing southeasterly through desert and semi-desert regions of Mongolia and Inner Mongolia before reaching Beijing. Each year dust storms carry particulate matter from the deserts of Gobi and Teklimakan towards Beijing, especially during the spring; other lithogenic sources of dust are bare soils, coal heaps and construction sites occurring in and around Beijing. The air masses associated with clusters 4 and 5 however, were from easterly and southerly directions respectively that have passed over regions with a variable vegetated cover.

On the map showing results of the Potential Source Contribution Function (PSCF) analysis (Fig. 3) high values of PSCF (>0.4) for PM_{10} are found along Bohai Bay and in Hebei and Shandong Provinces, where heavy industries are situated. Particles from these regions are mainly associated with transport paths recorded by clusters 4 and 5 (Fig. 2). Based on the US Environmental Protection Agency's (EPA) Models-3/Community Multi-scale Air Quality (CMAQ) model simulation over the Beijing region, Streets et al. (2007) estimated that about 34% of $PM_{2.5}$ on average at the Olympic Stadium site is attributable to sources outside Beijing. Correspondingly our clustering results suggest that particle transport from Hebei (21.05%), Inner Mongolia (23.03%) and Shandong (23.03%) provinces had a significant impact on PM_{10} levels in Beijing between June and October 2008 (Fig. 2). Hence the local control measures in Beijing were clearly insufficient for achieving air quality goals set for the Beijing Olympics.

3.2. Meteorology, particle concentration and magnetic susceptibility

Fig. 4 summarizes the record of temperature, relative humidity and PM_{10} mass concentration observations covering the time interval of the Olympic Games compared with the experimental results of the present study for magnetic susceptibility. The temperature (T) shows an inverse correlation with relative humidity (RH), while the RH values remains high through the main interval, the PM_{10} mass concentration decreases to a minimum in the "during" period (Fig.4a-b). Magnetic susceptibility (κ) from the FT district adjoins the Jing-Shen Highway (see Fig. 1) and displays some correspondence with the PM_{10} mass concentration in

Beijing dropping sharply in the “before” period and rising gently in the “after” period (Fig.4c). Magnetic minerals in aerosols could be derived from combustion processes related to industry, domestic heating or vehicles (Petrovský & Elwood, 1999). Therefore, magnetic parameters, notably magnetic susceptibility, are possible proxies to monitor the relative changes of atmospheric PM pollution in an area over time. The RH during the sampling periods in 2008 were almost at the same level as in 2009, but the PM_{10} mass concentration was obviously lower (Fig. 4d). So the reduction in PM_{10} and magnetic susceptibility is attributable to the source control measures implemented during the Olympic and Paralympic Games; these included removing approximately one-half of the vehicles (~1.5 million cars) off the roads in Beijing on alternate days under an even-odd license plate system, closing pollution-emitting factories, and slowing down construction activities. The measures implemented were substantial and extended far beyond Beijing: in the neighboring Tianjin municipality, in Hebei, Shanxi, and Shandong provinces, and in the Inner Mongolia Autonomous Region, polluting factories were closed and high-emission cars removed from roads (Stone, 2008).

In our previous study of street dust in the Beijing Olympic Park we observed that magnetic compositions in street dust decreased significantly during the interval of the Olympic Games enabling magnetic measurements to serve as a tool for rapidly and efficiently monitoring the impact of control measures (Qiao et al., 2011a). The temporal distribution of magnetic concentration parameters in street dust showed a similar trend to the PM variation in the Chaoyang district. Nevertheless, the χ value of street dust at some sampling sites during the Olympic period was actually higher than the value at other times indicating a primary component of street dust originating mainly from sources in the immediate proximity.

3.3. Comparison of the elemental composition of street dust and PM samples

The heavy metal concentration values determined from inductively-coupled plasma-mass spectrometry are listed in Table 2. The order for the mean concentrations of eight heavy metals in the PM is Zn>Cr>Pb>Cu>V>Ni>Fe>Co; zinc and cobalt remain the most and least abundant metals respectively in the street dust but the intervening order is somewhat different: Zn>V>Cr>Cu>Pb>Fe>Ni>Co. The relative importance of these elements, particularly the consistent relatively-high amounts of Zn, seems to have no straightforward explanation. Unfortunately vehicle emission investigations focus on the gas content and group particulate matter is quoted without constituent metal analysis. To facilitate a comparison, normalized elemental compositions of street dust and PM samples are compared in Table 2 where elemental mass ratios have been normalized with respect to the (mainly crustal-derived) elemental aluminum for clarity. As shown in Table 2, except for Cr and Pb the mean and normalized concentrations of other heavy metals for street dust exceed the PM values, especially the Zn. In part, the elemental concentrations at a specific location will be determined by the distance from their sources and reflect emissions from point-sources emitters. The FT site (Fig. 1) located close to the Jing-Shen Highway was once the highest polluting industrial area of Beijing where Zn and perhaps Pb are more likely to have been retained by refining processes in contrast to the other elements found here in greater abundance. Metallurgical processes have been found to produce the largest emissions of Cu, Ni and Zn (Pacyna, 1998), while exhaust emissions from road vehicles also contain various amounts of these metals (Pacyna, 1986; Lee et al., 1999). Johansson et al. (2009) found that vehicle emissions with high loadings of Cu and Zn were an important source of street dust. The mobility of each element will also have a part to play here: with voluminous vehicle and industrial emissions of polluting elements such as Zn and Pb, it is unsurprising that the results identify higher concentrations of these elements compared with the largely-residential area SLT to the northwest. The mass concentrations of all 8 elements (Fe, V, Cr, Co, Ni, Cu, Zn and Pb) for street dust in the traffic site are higher than those in the park site and a primary origin in vehicular source emission is obvious; this is especially the case for Zn indicating that vehicle exhaust is likely an important source of heavy metals in the Chaoyang District, although the full balance of sources responsible for the high Zn is presently unclear.

Since leaded gasoline was phased out in Beijing in 1997, motor exhaust emissions seem unlikely to be the dominant source of Pb in the aerosols unless they result from erosion of engines and exhausts. Liu et al. (2005) suggest that when winds from southern and southwestern directions prevail in Beijing, high concentrations of vapor and air pollutants from power plants, refining industries and biomass burning enhance the aerosol concentrations of Pb and Zn (found together in primary ores). Re-suspended dust could be the possible sources of these metals and an additional factor here is the construction and demolition activity before the Olympic Games resulting in construction dust being an important contributor to PM and street dust prior to commencement of the Games.

The I_{geo} values for heavy metals in the PM and street dust from Chaoyang District are listed in Table 3 and plotted in Fig. 5. The Geo-accumulation index results reveal PM contents at the SLT and FT, traffic and park sites where street dust is uncontaminated to only moderately contaminated with respect to Cu, Zn and Pb although the I_{geo} value for Zn in the traffic area are especially high and classified as extremely contaminated (Table 3). In contrast the results of I_{geo} in the park area indicate little or no metal contamination.

The average I_{geo} values for heavy metals in PM are -1.15 for Fe, -1.46 for V, -0.39 for Cr, -1.80 for Co, -0.76 for Ni, 0.44 for Cu, 0.96 for Zn and 1.45 for Pb respectively. The maximum I_{geo} values for V, Fe and Co are less than zero, indicating that PM in the study areas is contaminated by other metals. The maximum I_{geo} values for V, Cr, Fe, Co and Ni in street dust are less than zero and ranked as "uncontaminated". The I_{geo} values for the other metals such as Cu, Zn and Pb are all greater than zero, indicating that street dust in Chaoyang District is polluted by these metals to varying degrees (Fig.5b). The same phenomena is reported for road dusts from Bulgaria and Greece (Bourliva et al., 2016; Jordanova et al., 2014). Zn in the traffic area is recognized as extremely contaminated (c.f. Müller, 1979) whilst in the park area I_{geo} values are less than zero. As in the previous research (Qiao et al., 2011a) the high Zn concentrations in traffic areas is attributable to vehicle emissions, and is probably ultimately sourced in corrosion of the vehicle engines and body work. I_{geo}

values for Cu, Zn and Pb are higher than zero for both PM and street dust samples indicating that dust in the Chaoyang District is contaminated by the metals derived from anthropogenic sources: in contrast the metals V, Fe and Co are evidently not readily shifted into the environment by engine usage. The high Pb fluxes associated with past emissions likely stored in soils that are now being remobilized by surface erosion processes to contribute to PM contamination.

4. Conclusions

Five atmospheric trajectories to aerosols arriving in Beijing during the period from June 2008 to October 2008 spanning the interval of the 2008 Olympic Games were identified for application of cluster analysis methodology. Pathways associated with trajectory clusters 4 and 5 passing over Shandong and Hebei provinces before reaching Beijing account for 23.03% and 21.05% of all PM. PSCF analysis also identifies high PM values in these provinces as well as along Bohai Bay. Dust particles in Beijing came mainly from distant transport from the south and southeast and were then supplemented by local emissions.

The study clearly shows that levels of PM_{10} and κ were lower during the Olympic period compared with the pre-Olympic period, and there is no doubt that the measures planned to limit air pollution in Beijing greatly improved air quality during the interval of the 2008 Olympic Games although the limited post-Olympic record suggests that major improvement is unlikely to be sustained without the imposition of more restrictive measures. PM and street dust remains heavily contaminated with Cu, Zn and Pb and this is prominently the case for Zn where I_{geo} attains a value of 3.5 in the street dust. Even with firm and widespread controls the levels of heavy metal pollution remain high for historic reasons and imply that the limit of zero emissions cannot be achieved in practice. Furthermore, the I_{geo} values for the street dust are generally higher than those for PM of a given element and district. Thus, street dust tends to be more heavily contaminated than the PM. The distribution of the heavy metal concentration in street dust in the study area indicates that vehicle emissions are mainly responsible for heavy metal pollution as shown by the highest heavy metal concentrations found in the traffic site.

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REFERENCES

Almeida, S.M., Pio, C.A., Freitas, M.C., Reis, M.A., Trancoso, M.A., 2006. Source apportionment of atmospheric urban aerosol based on weekdays/weekend variability: evaluation of road resuspended dust contribution. Atmos. Environ. 40, 2058-2067.

Amato, F., Pandolfi, M., Escrig, A., Querol, X., Alastuey, A., Pey, J., Perez, N., Hopke, P.K., 2009. Quantifying road dust resuspension in urban environment by Multilinear Engine: A comparison with PMF2. Atmos. Environ. 43(17), 2770-2780.

Ashbaugh, L.L., 1983. A statistical trajectory technique for determining air pollution source regions. J. Air Pollut. Control Assoc. 33, 1096-1098.

Borge, R., Lumbreras, J., Vardoulakis, S., Kassomenos, P., Rodríguez, E., 2007. Analysis of long-range transport influences on urban PM_{10} using two-stage atmospheric trajectory clusters. Atmos. Environ. 41, 4434-4450.

Bourliva, A., Papadopoulou, L., Aidona, E., 2016. Study of road dust magnetic phases as the main carrier of potentially harmful trace elements. Sci. Total Environ.553, 380-391.

China National Environmental Monitoring Center (CNEMC), 1990. The background values of elements in Chinese soils. China Environmental Science Press, Beijing (in Chinese).

Cinzia Perrino, 2010. Atmospheric particulate matter. Proceedings of a C.I.S.B. Minisymposium, 35-43.

Forstner, U., Ahlf, W., Calmano, W., Kersten, M., 1990. Sediment criteria development- contributions from environmental geochemistry to water quality management, in: Heling D. et al. (Eds.), Sediments and environmental geochemistry: selected aspects and case histories. Berlin, pp. 311-338.

Georgeaud, V.M., Rochette, P., Ambrosi, J.P., Vandamme, D., Williamson, D., 1997. Relationship between heavy metals and magnetic properties in a large polluted catchment: The Etang de Berre (south of France). Phys. Chem. Earth 22, 211-214.

Harrison, R.M., Laxen, D.P.H., Wilson, S.J., 1981. Chemical associations of Lead, Cadmium, Copper and Zinc in street dusts and roadside soils. Environ. Sci. Technol. 15, 1378-1383.

Hien, P.D., Binh, N.T., Truong, Y., Ngo, N.T., 1999. Temporal variations of source impacts at the receptor, as derived from air particulate monitoring data in Ho Chi Minh City, Vietnam. Atmos. Environ. 33, 3133-3142.

Johansson, C., Norman, M., Burman, L., 2009. Road traffic emission factors for heavy metals. Atmos. Environ. 43, 4681-4688.

Jordanova, D., Jordanova, N., Petrov, P., 2014. Magnetic susceptibility of road deposited sediments at a national scale-Relation to population size and urban pollution. Env. Poll. 189, 239-251.

Lee, E., Chan, C.K., Paatero, P., 1999. Application of positive matrix factorization in source apportionment of particulate pollutants in Hong Kong. Atmos. Environ. 33, 3201-3212.

Li, Y., Shao, M., Lu, S.H., Chang, C.C., Dasgupta, P.K., 2010. Variations and sources of ambient formaldehyde for the 2008 Beijing Olympic games. Atmos. Environ. 44, 2632-2639.

Liu, Q.S., Roberts, A.P., Larrasoaña, J.C., Banerjee, S.K., Guyodo, Y., Tauxe, L., Oldfield, F., 2012. Environmental magnetism: principles and applications. Rev. Geophys. 50, doi: 10.1029/2012RG000393.

Maher, B.A., Thompson, R., Hounslow, M.W., 1999. Magnetic monitoring of air-land- and water-pollution, in: Maher, B.A., Thompson, R. (Eds.), Quaternary Climates, Environments and Magnetism. Cambridge University Press, New York, pp.1-48.

Martuzevicius, D., Kliucininkas, L., Prasauskas, T., Krugly, E., Kauneliene, V., Strandberg, B., 2011. Resuspension of particulate matter and PAHs from street dust. Atmos. Environ. 45(2), 310-317.

Morris, M.A., Versteeg, J.K., Bryant, D.W., Legzdins, A.E., McCarry, B.E., Marvin, C.H., 1995. Preliminary comparisons between mutagenicity and magnetic susceptibility of respirable airborne particulate. Atmos. Environ. 29, 3441-3450.

Müller, G., 1969. Index of geoaccumulation in sediments of the RhineRiver. Geojournal 2, 108-118.

Müller, G., 1979. Heavy metals in the sediment of the Rhine-Changes Seity. Umsch. Wiss. Tech. 79, 778-783.

Pacyna, J.M., 1986. Atmospheric trace elements from natural and anthropogenic sources, In: Nriagu, J.O., Davidson, C.I. (Eds.), Toxic metals in the atmosphere. John Wiley & Sons, New York, pp. 33-52.

Pacyna, J.M., 1998. Source inventories for atmospheric trace metals, In: Harrison, R.M., van Grieken, R.E. (Eds.), Atmospheric particles, IUPAC series on analytical physical chemistry of environmental systems. Wiley, Chichester, UK, pp. 385-423.

Petrovský, E., Elwood, B., 1999. Magnetic monitoring of air-, land- and water pollution, In: Maher, B., Thompson, R. (Eds.), Quaternary Climates, Environments and Magnetism. Cambridge University Press, Cambridge, pp. 279-322.

Qiao, Q.Q., Huang, B.C., Zhang, C.X., John D.A. Piper, Pan, Y.P., Sun, Y., 2013. Assessment of heavy metal contamination of dustfall in northern China from integrated chemical and magnetic investigation. Atmos. Environ. 74, 182-193.

Qiao, Q.Q., Zhang, C.X., Huang, B.C., John D.A. Piper, 2011a. Evaluating the environmental quality impact of the 2008 Beijing Olympic Games: magnetic monitoring of street dust in Beijing Olympic Park. Geophys. J. Int. 187, 1222-1236.

Qiao, Q.Q., Zhang, C.X., Li, J., Li, H., Huang, B.C., 2011b. Magnetic properties and indicator of concentration of pollution of atmospheric dust in Chaoyang, Beijing. Chinese J. Geophys. 54, 151-162 (in Chinese).

Rolph, G.D., 2016. Real-time environmental applications and display system (READY) website (http://www.ready.noaa.gov). NOAA Air Resources Laboratory, College Park, MD.

Sirois, A., Bottenheim, J.W., 1995. Use of backward trajectories to interpret the 5-year record of PAN and O_3 ambient air concentrations at Kejimkujik National Park, Nova Scotia. J. Geophys. Res. 100, 2867-2881.

Stein, A.F., Draxler, R.R., Rolph, G.D., Stunder, B.J.B., Cohen, M.D., Ngan, F., 2015. NOAA's HYSPLIT atmospheric transport and dispersion modeling system. Bull. Amer. Meteror. Soc. 96, 2059-2077.

Stone, R., 2008. Beijing's marathon run to clean foul air nears finish line. Science, 321, 636-637.

Streets, D.G., Fu, J.S., Jang, C.J., Hao, J.M., He, K.B., Tang, X.Y., Zhang, Y.H., Wang, Z.F., Li, Z.P., Zhang, Q, Wang L.T., Wang, B.Y. and Yu, C., 2007. Air quality during the 2008 Beijing Olympic Games. Atmos. Environ. 41, 480-492.

Thornton, I., 1991. Metal contamination of soils in urban areas, in: Bullock, P., Gregory, P.J. (Eds.), Soils in the Urban Environment. Blackwell, UK, pp.47-75.

Wang, Y.Q., Zhang, X.Y., Arimoto, R., 2006. The contribution from distant dust sources to the atmospheric particulate matter loadings at XiAn, China during spring. Sci. Total Environ. 368, 875-883.

Wang, Y.Q., Zhang, X.Y., Draxler, R.R., 2009. TrajStat: GIS-based software that uses various trajectory statistical analysis methods to identify potential sources from long-term air pollution measurement data. Environ. Model. Softw. 24, 938-939.

Wise, E.K., Comrie, A.C., 2005. Meteorologically adjusted urban air quality trends in the Southwestern United States. Atmos. Environ. 39, 2969-2980.

Zhao, H.T., Shao, Y.P., Yin, C.Q., Jiang, Y., Li, X.Y., 2016. An index for estimating the potential metal pollution contribution to atmospheric particulate matter from road dust in Beijing. Sci. Total Environ. 550, 167-175.

Zhou, Y., Wu, Y., Yang, L., Fu, L.X., He, K.B., Wang, S.X., Hao, J.M., Chen, J.C., Li, C.Y., 2010. The impact of transportation control measures on emission reductions during the 2008 Olympic Games in Beijing, China. Atmos. Environ. 44, 285-293.

Figure Captions:

Figure 1: Schematic map of the study area showing sampling locations of atmospheric PM and street dust.

Figure 2: Cluster-mean back trajectories for Beijing during the period covered by the Olympic Games (June 2008 and October 2008).

Figure 3: Potential source contribution function (PSCF) map for Beijing PM_{10} for the interval between June 2008 and October 2008.

Figure 4: Time series of (a) temperature and relative humidity (RH) between June 2008 and October 2008. (b) Relative humidity and PM_{10} mass concentration also between June 2008 and October 2008. (c) PM_{10} mass concentration variation and smoothed magnetic susceptibility (κ) covering the same interval between June 2008 and October 2008. (d) PM_{10} mass concentration and relative humidity (RH) in 2008 and 2009 covering the broader interval before, during and after the period of Olympic Games.

Figure 5: Geo-accumulation index (I_{geo}) for elements (a) in dusts of PM compared with (b) street dust from the Chaoyang District.

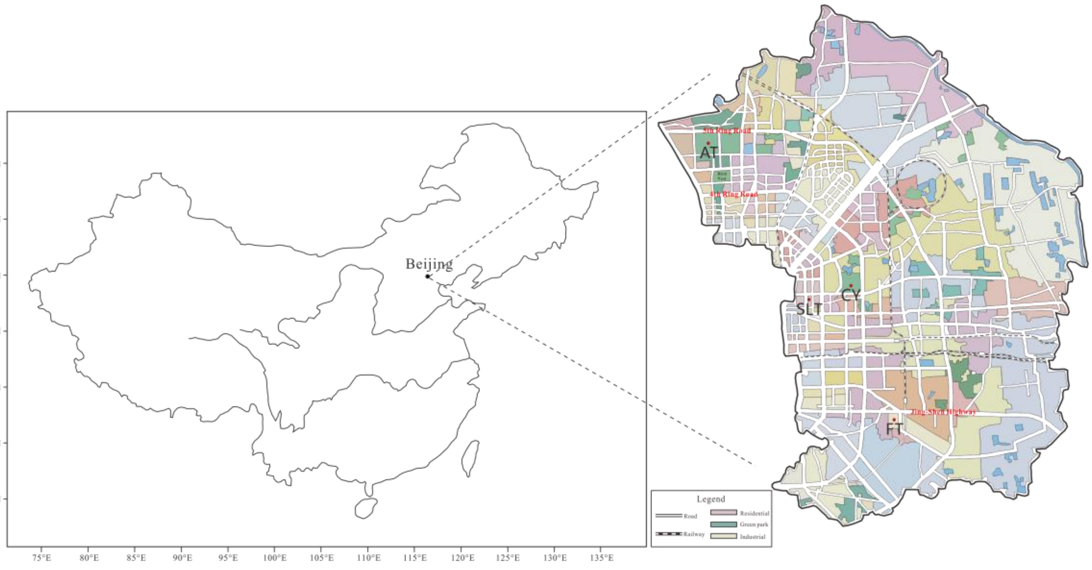


Figure 1

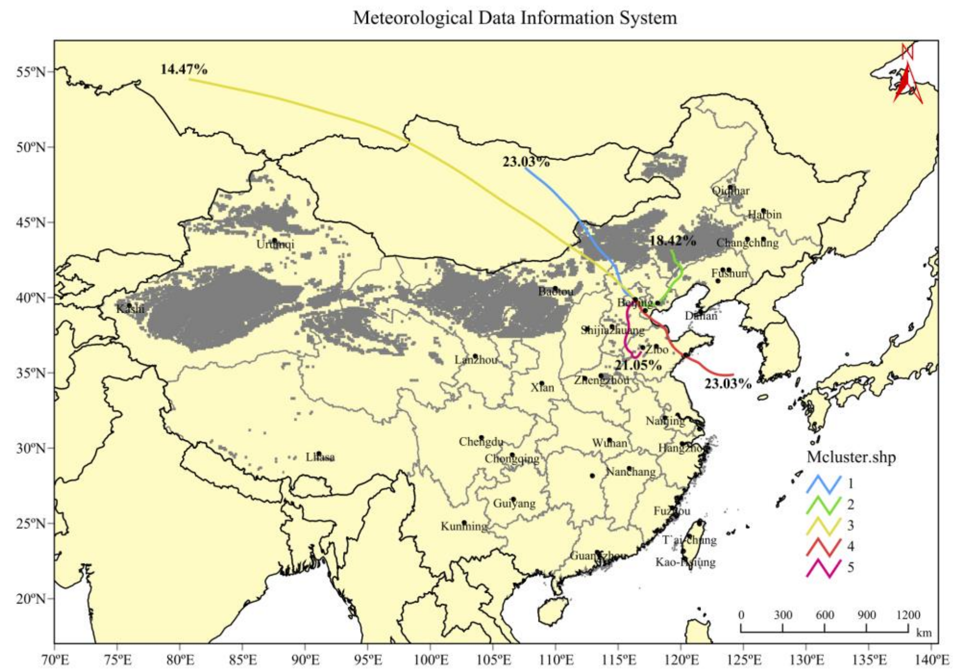
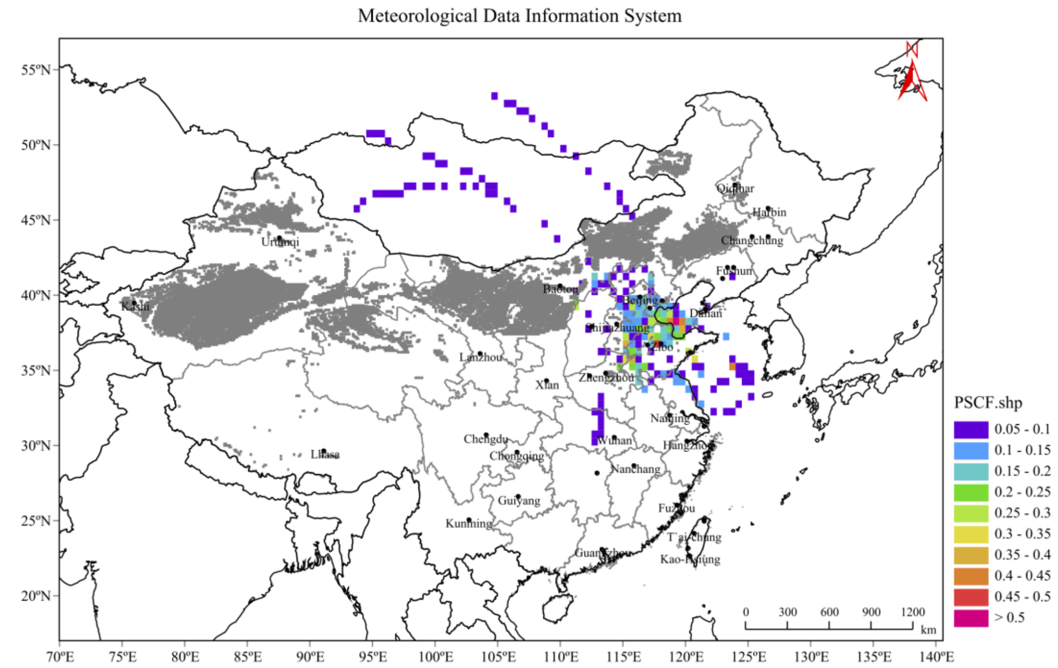


Figure 2



Meteorological Data Information System
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130ºE
135ºE
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Qiqihar
Harbin
Changchung
Urumqi
Fushun
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Kashi
Dalian
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Zibo
Lanzhou
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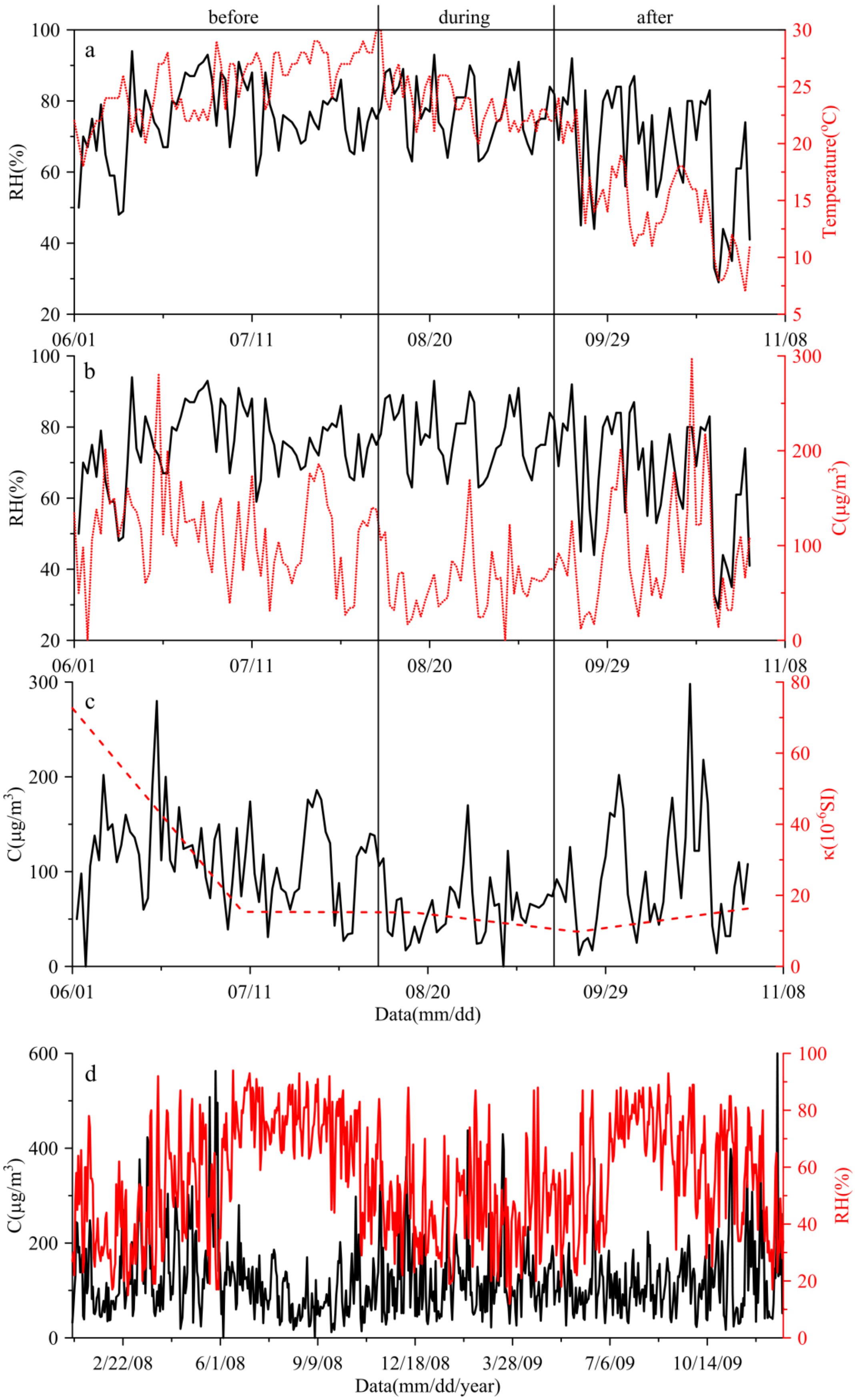
Nanjing
Chengdu
Wuhan
Hangzhou
Chongqing
Lhasa
Nanchang
Guiyang
Fuzhou
Kunming
T'aichung
Guangzhou
Kao-hsiung
0 300 600 900 1200
km
PSCF.shp
0.05 - 0.1
0.1 - 0.15
0.15 - 0.2
0.2 - 0.25
0.25 - 0.3
0.3 - 0.35
0.35 - 0.4
0.4 - 0.45
0.45 - 0.5
> 0.5

Figure 3

466



467 Figure 4

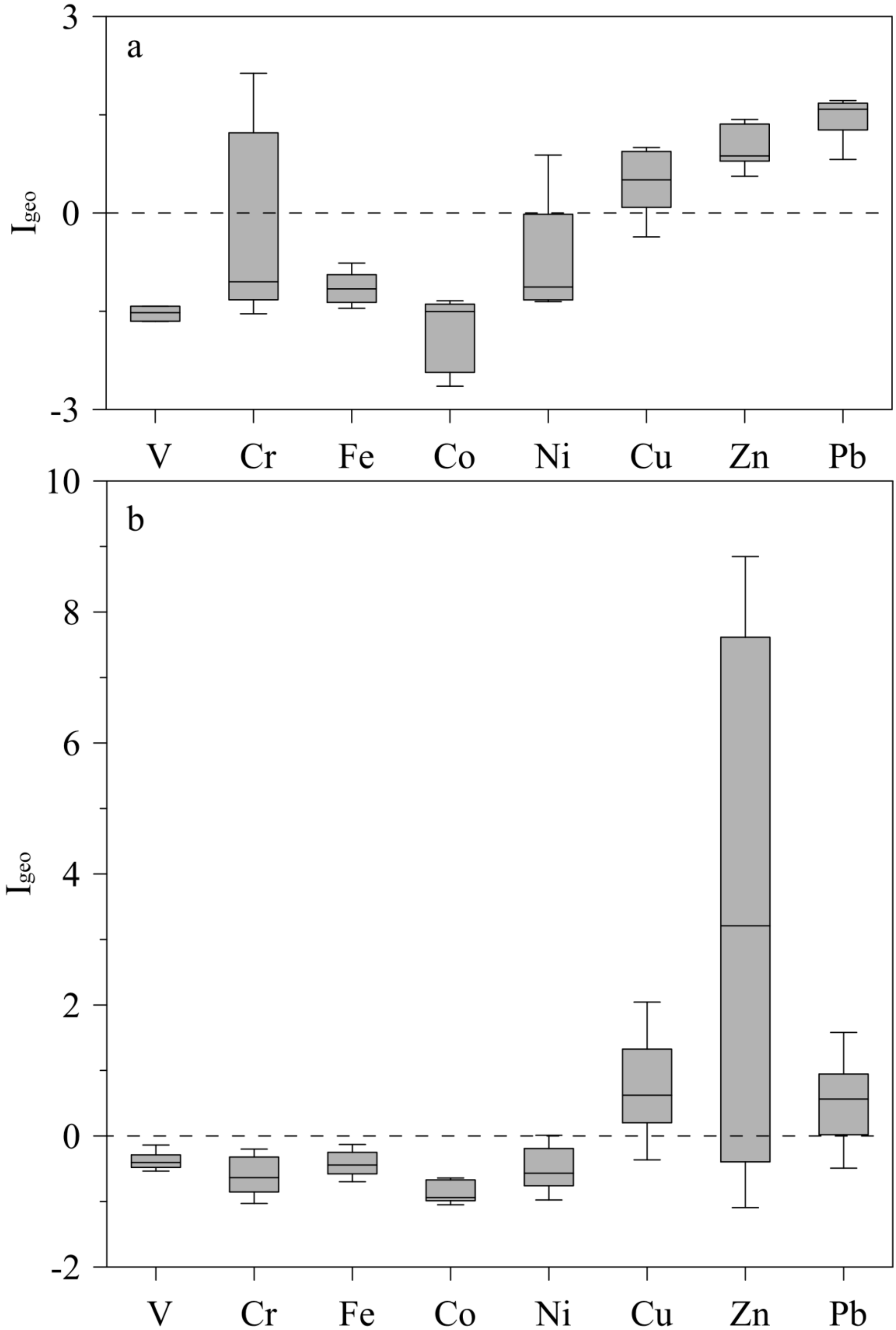


Figure 5

Table 1

The seven classes comprising the Geo-accumulation index.

Class	Value	Particulate matter quality
0	$I_{geo} \leq 0$	Practically uncontaminated
1	$0 < I_{geo} < 1$	Uncontaminated to moderately contaminated
2	$1 < I_{geo} < 2$	Moderately contaminated
3	$2 < I_{geo} < 3$	Moderately to heavily contaminated
4	$3 < I_{geo} < 4$	Heavily contaminated
5	$4 < I_{geo} < 5$	Heavily to extremely contaminated
6	$5 < I_{geo}$	Extremely contaminated

Table 2 Summary of the heavy metal content, normalized elemental compositions and magnetic susceptibility of atmospheric PM and street dust samples collected in the Chaoyang District. Values are given in mg/kg except for Fe where values are in g/kg. Magnetic susceptibility values are denoted as κ for PM and χ for street dust.

Site ID		**Fe**	**V**	**Cr**	**Co**	**Ni**	**Cu**	**Zn**	**Pb**	**magnetic susceptibility**
	Max	26.21	61.32	447.95	8.91	80.28	70.78	283.52	104.55	28.24
SLT(n=3)	Min	19.96	40.21	40.78	3.74	17.01	27.48	226.93	67.16	4.07
	Mean±SD	**23.12±3.12**	**48.59±11.20**	**179.36±232.65**	**7.08±2.89**	**38.24±36.40**	**46.78±22.03**	**263.94±32.07**	**87.82±19.01**	**12.17**
	normalized	**0.29**	**0.61**	**2.25**	**0.09**	**0.48**	**0.59**	**3.32**	**1.1**	
	Max	20.29	43.27	238.66	9.24	42.91	67.87	414.03	125.21	72.65
FT(n=4)	Min	16.24	37.69	35.17	4.33	17.33	37.57	266.42	114.26	9.75
	Mean±SD	**18.30±1.86**	**40.02±2.75**	**92.63±97.61**	**7.08±2.15**	**25.25±11.87**	**53.02±12.62**	**336.90±78.07**	**119.67±4.79**	**25.85**
	normalized	**0.23**	**0.50**	**1.16**	**0.09**	**0.32**	**0.67**	**4.23**	**1.50**	
	Max	26.21	61.32	447.95	9.24	80.28	70.78	414.03	125.21	72.65
PM(n=7)	Min	16.24	37.69	35.17	3.74	17.01	27.48	226.93	67.16	4.07
	Mean±SD	**20.36±3.41**	**43.70±8.16**	**129.8±157.97**	**7.08±2.26**	**30.82±23.67**	**50.35±15.89**	**305.63±70.08**	**106.02±20.54**	**19.01**
	normalized	**0.26**	**0.55**	**1.63**	**0.09**	**0.39**	**0.63**	**3.84**	**1.33**	
	Max	40.70	108.00	88.90	15.00	43.80	146.00	70828.00	114.00	495.98
Traffic(n=5)	Min	34.36	90.00	73.40	12.30	31.30	63.20	15106.00	68.10	284.24
	Mean±SD	**37.27±2.35**	**98.00±7.05**	**79.65±6.01**	**14.13±0.98**	**36.67±4.48**	**97.13±32.27**	**32968.17±19695.27**	**79.30±17.26**	**391.05**
	normalized	**0.47**	**1.23**	**1.00**	**0.18**	**0.46**	**1.22**	**414.17**	**0.99**	
	Max	31.25	89.10	58.80	12.10	27.50	47.00	134.00	46.60	72.76
Park(n=5)	Min	27.44	81.90	50.00	11.30	22.10	27.50	72.10	27.10	25.65
	Mean±SD	**29.43±1.33**	**84.78±2.77**	**54.90±3.06**	**11.73±0.29**	**25.25±1.82**	**36.02±7.98**	**106.27±24.10**	**38.70±7.16**	**56.94**
	normalized	**0.37**	**1.07**	**0.69**	**0.15**	**0.32**	**0.45**	**1.34**	**0.49**	
	Max	40.70	108.00	88.90	15.00	43.80	146.00	70828.00	114.00	495.98
Dust(n=10)	Min	27.44	81.90	50.00	11.30	22.10	27.50	72.10	27.10	25.65
	Mean±SD	**33.35±4.48**	**91.39±8.58**	**67.28±13.70**	**12.93±1.43**	**30.96±6.79**	**66.58±38.99**	**16537.22±21698.84**	**59.00±24.66**	**247.86**
	normalized	**0.42**	**1.15**	**0.85**	**0.16**	**0.39**	**0.84**	**207.75**	**0.74**	

normalized mass concentration of element species in PM and street dust, relative to Al

The mean values for SLT, FT, traffic and park are means of monthly values at one location; the mean values for PM and dust are means of monthly values at different locations

Table 3 Geo-accumulation indices for elements in PM and street dust

Site ID	**Fe**	**V**	**Cr**	**Co**	**Ni**	**Cu**	**Zn**	**Pb**
SLT(n=3)	-0.96	-1.31	-0.08	-1.78	-0.60	0.29	0.77	1.18
FT(n=4)	-1.29	-1.57	-0.62	-1.83	-0.88	0.55	1.10	1.65
PM(n=7)	-1.15	-1.46	-0.39	-1.80	-0.76	0.44	0.96	1.45
Traffic(n=5)	-0.26	-0.28	-0.36	-0.73	-0.26	1.39	7.57	1.03
Park(n=5)	-0.60	-0.49	-0.90	-0.99	-0.79	-0.004	-0.57	0.0002
Dust(n=10)	-0.43	-0.38	-0.63	-0.86	-0.52	0.69	3.50	0.52